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Compressed Liquid Densities of Methyl *tert*-Butyl Ether (MTBE), Ethyl *tert*-Butyl Ether (ETBE), and Diisopropyl Ether (DIPE)

E. Christian Ihmels and Jürgen Gmehling*

Carl von Ossietzky University of Oldenburg, Department of Industrial Chemistry, P.O. Box 2503, D-26111 Oldenburg, Germany

Densities of methyl *tert*-butyl ether (MTBE), ethyl *tert*-butyl ether (ETBE), and diisopropyl ether (DIPE) have been measured with a computer-controlled high-temperature, high-pressure vibrating tube densimeter system (DMA-HDT) in the liquid state. The uncertainty in the density measurement was estimated to be lower than ±0.2 $kg \cdot m^{-3}$. The densities were measured for temperatures from 273 K to 473 K and pressures up to 40 MPa for MTBE (337 data points) and for ETBE (364 data points) and up to 35 MPa for DIPE (328 data points), which covers the density range between (459 and 792) $kg \cdot m^{-3}$ for MTBE, between (494 and 792) $kg \cdot m^{-3}$ for ETBE, and between (471 and 774) $kg \cdot m^{-3}$ for DIPE. The experimental data were correlated with the new three-dimensional density correlation system (TRIDEN) and compared with published data.

Introduction

During the last two decades, branched alkyl ethers have become important as fuel additives. The ethers increase the amount of oxygen in gasoline in order to reduce the CO content in the exhaust gas of automobiles, and they are a substitute for tetraethyl lead, which was formerly used to increase the octane number. With a usage of more than 30 000 tons per day in the United States, methyl *tert*-butyl ether (MTBE) is a large scale product and the main oxygenated compound currently used. Because of the high water solubility and the health risk caused by the contamination of drinking water, MTBE has become a very controversial gasoline additive. The isomer ethers ethyl *tert*-butyl ether (ETBE) and diisopropyl ether (DIPE) have some important advantages compared to MTBE, for example, being chemically more similar to hydrocarbons and having a lower solubility in water.[1] Finally, the higher boiling points of ETBE and DIPE allow incorporation of more light feedstocks in gasoline, especially light cyclic alkanes which are also used as octane enhancers.[2]

The compressed liquid densities of MTBE, ETBE, and DIPE have been measured with a computer-controlled vibrating tube densimeter. The densimeter had been calibrated with degassed water and butane and by vacuum measurements. Densities were measured for temperatures from 273 K to 473 K and pressures up to 40 MPa for MTBE (337 data points) and for ETBE (364 data points) and up to 35 MPa for DIPE (328 data points), which covers the density range between (459 and 792) $kg \cdot m^{-3}$ for MTBE, between (494 and 792) $kg \cdot m^{-3}$ for ETBE, and between (471 and 774) $kg \cdot m^{-3}$ for DIPE. The measured densities of MTBE, ETBE, and DIPE were correlated with the three-dimensional correlating model TRIDEN. Details of the measurement system, the calibration, and the correlation can be found in refs 3 and 4.

* To whom correspondence should be addressed. E-mail: gmehling@tech.chem.uni-oldenburg.de. Telephone: ++49-441-798-3831. Fax: ++49-441-798-3330.

Experimental Section

Methyl *tert*-butyl ether (MTBE, $C_5H_{12}O$, M = 88.15 $g \cdot mol^{-1}$, CAS-RN 1634-04-4), ethyl *tert*-butyl ether (ETBE, $C_6H_{14}O$, M = 102.18 $g \cdot mol^{-1}$, CAS-RN 637-92-3), and diisopropyl ether (DIPE, $C_6H_{14}O$, M = 102.18 $g \cdot mol^{-1}$, CAS-RN 108-20-3) were obtained by Büfa Oldenburg (MTBE), Veba Oel (ETBE), and Aldrich (DIPE). All compounds were stored over 3 Å molecular sieves and degassed by distillation. The final purities were checked by gas chromatography (MTBE, >99.9 mass %; ETBE, >99.5 mass %; DIPE, >99.8 mass %) and by Karl Fischer titration (water content for all compounds: <10 ppm).

A computer-operated vibrating tube densimeter system for high temperatures and pressures (temperatures from 273 K to 623 K and pressures up to 40 MPa) was used for the density measurements. The automated equipment can be used for the determination of densities in the sub- and supercritical states. With this apparatus, a large number of data points can be obtained in a rather short time with a minimum of manual effort. A temperature and pressure program can be driven to obtain a complete $P\rho T$ surface for the desired component. The measurement system was developed in the thesis of Ihmels.[5] Densities of several liquids and liquefied gases (e.g. toluene, carbon dioxide, carbonyl sulfide, hydrogen sulfide, sulfur hexafluoride, dinitrogen monoxide, and R227ea) have already been published.[3−6] The comparison with reference equations of state (EoS) for toluene, CO_2, and SF_6 demonstrated the high accuracy and suitability of this measurement system.

The apparatus and procedure of the measurements were described in detail by Ihmels and Gmehling.[3,4] A scheme of the density measurement system is shown in Figure 1. A prototype of a high-pressure high-temperature vibrating tube densimeter (DMA-HDT) is the essential part of the experimental setup. The temperature is measured using a Pt100 resistance thermometer, and the pressure is monitored by means of a calibrated external pressure sensor (model PDCR 911, pressure range 60 MPa, Druck). The density values are obtained from the periods of oscillation of the vibrating tube. For the calibration, the period of

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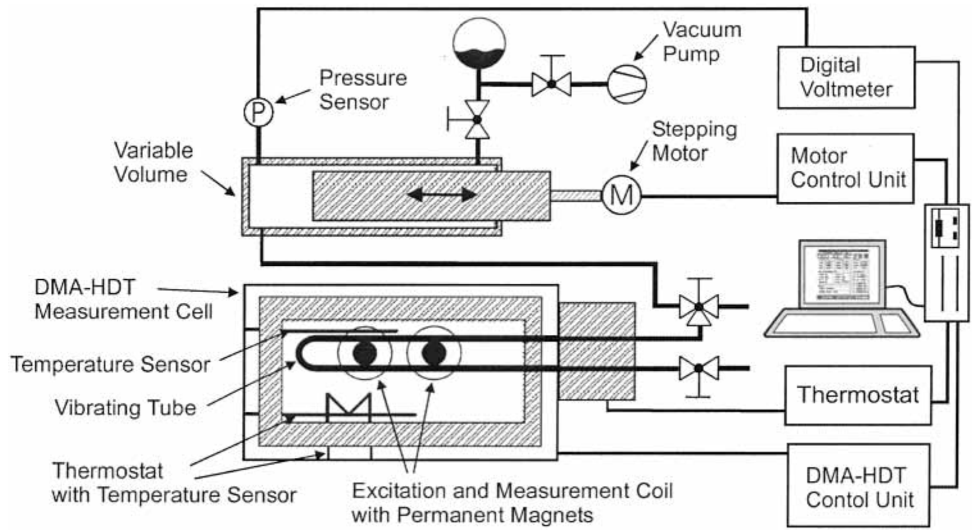

Figure 1. Schematic diagram of the computer-controlled density measurement unit.

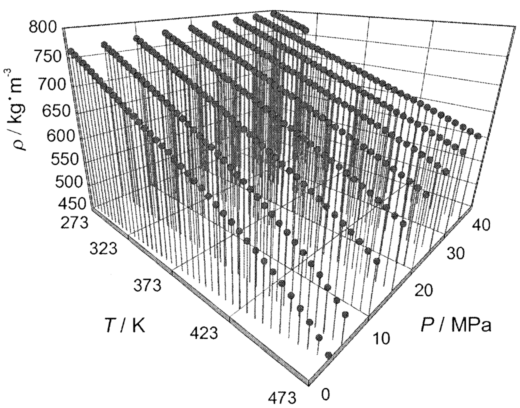

Figure 2. Densities of methyl *tert*-butyl ether (MTBE) at temperatures between 273 K and 473 K and pressures between 0.3 and 40 MPa.

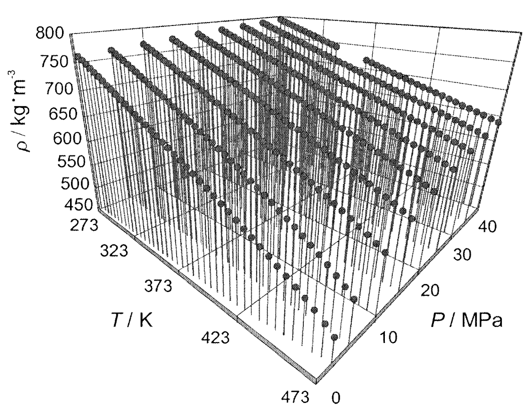

Figure 3. Densities of ethyl *tert*-butyl ether (ETBE) at temperatures between 273 K and 473 K and pressures between 0.3 and 40 MPa.

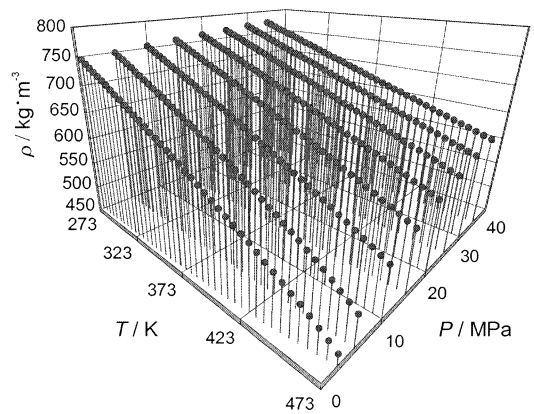

Figure 4. Densities of diisopropyl ether (DIPE) at temperatures between 273 K and 473 K and pressures between 0.3 and 35 MPa.

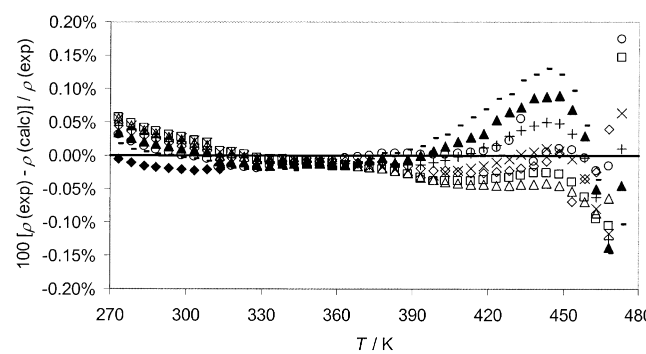

Figure 5. Relative deviations between experimental densities and the TRIDEN correlation for methyl *tert*-butyl ether (MTBE): ○, <5 MPa; ◇, 5 MPa; △, 10 MPa; □, 15 MPa; ×, 20 MPa; +, 25 MPa; ▲, 30 MPa; −, 35 MPa; ◆, 40 MPa.

oscillation at zero pressure and the two reference substances water and butane were used. The reference densities were calculated using the reference EoS from Pruβ and Wagner[7,8] for water and the EoS from Younglove and Ely[9] for butane. The uncertainty of the temperature is estimated to be ±0.03 K, and the pressure has an estimated uncertainty of ±6 kPa. For density measurements in the temperature and pressure ranges covered (273 K to 473 K and 0.3 MPa to 40 MPa) a maximum error of ±0.2 kg·m^{-3} is estimated.

Results and Discussion

In this work, the densities of MTBE (337 data points), ETBE (364 data points), and DIPE (328 data points) in the compressed liquid state were measured from 273 K to 473 K at 5 K intervals and from 0.3 MPa up to 40 MPa (for DIPE up to 35 MPa) at 5 MPa intervals. The results are listed in Tables 1−3 and presented graphically in Figures 2−4.

Besides the density measurements at atmospheric or saturation pressure published[10] for MTBE, ETBE, and DIPE up to 333 K, compressed liquid densities have already

Table 1. Experimental Densities of Methyl *tert*-Butyl Ether (MTBE)

T	P	ρ	T	P	ρ	T	P	ρ	T	P	ρ	T	P	ρ	T	P	ρ
K	MPa	$kg \cdot m^{-3}$	K	MPa	$kg \cdot m^{-3}$	K	MPa	$kg \cdot m^{-3}$	K	MPa	$kg \cdot m^{-3}$	K	MPa	$kg \cdot m^{-3}$	K	MPa	$kg \cdot m^{-3}$
273.21	0.318	761.07	303.22	14.976	746.83	338.15	0.486	691.94	373.14	0.707	649.88	408.13	1.131	601.65	443.13	1.840	541.68
273.21	4.970	765.60	303.22	19.980	751.70	338.15	4.961	699.54	373.14	4.964	660.29	408.13	4.970	616.35	443.13	4.953	563.86
273.21	9.973	770.15	303.22	24.970	756.23	338.15	9.977	707.13	373.14	9.969	670.70	408.13	9.964	631.36	443.13	9.967	587.54
273.20	14.977	774.40	303.22	29.975	760.48	338.15	14.984	713.95	373.14	14.961	679.69	408.13	14.958	643.57	443.13	14.982	604.80
273.20	19.967	778.37	303.22	34.970	764.45	338.15	19.962	720.13	373.14	19.960	687.67	408.13	19.970	654.00	443.13	19.988	618.61
273.20	24.982	782.10	303.22	39.956	768.18	338.15	24.970	725.82	373.14	24.977	694.87	408.13	24.982	663.12	443.13	24.964	630.15
273.21	29.955	785.62	308.22	0.397	724.88	338.15	29.962	731.08	373.14	29.973	701.41	408.13	29.971	671.22	443.13	29.985	640.25
273.20	34.974	788.95	308.22	4.971	730.81	338.15	34.973	735.98	373.14	34.970	707.39	408.13	34.963	678.54	443.13	34.968	649.16
273.20	39.954	792.07	308.22	9.975	736.74	343.15	0.509	686.21	378.14	0.751	643.45	413.13	1.216	594.04	448.13	1.969	531.33
278.20	0.359	756.03	308.22	14.966	742.17	343.15	4.977	694.17	378.14	4.977	654.40	413.13	4.985	609.58	448.13	4.956	555.18
278.20	4.953	760.69	308.22	19.966	747.22	343.15	9.958	702.04	378.14	9.982	665.32	413.13	9.963	625.43	448.13	9.968	580.68
278.21	9.967	765.42	308.22	24.968	751.90	343.15	14.981	709.15	378.14	14.969	674.69	413.13	14.965	638.24	448.13	14.971	598.85
278.21	14.973	769.82	308.22	29.978	756.29	343.15	19.984	715.58	378.14	19.979	682.99	413.13	19.990	649.12	448.13	19.982	613.27
278.21	19.969	773.93	308.22	34.980	760.38	343.15	24.971	721.44	378.14	24.970	690.40	413.13	24.976	658.50	448.13	24.970	625.26
278.20	24.972	777.80	308.22	39.927	764.21	343.15	29.964	726.87	378.14	29.978	697.15	413.13	29.974	666.85	448.12	29.968	635.66
278.21	29.969	781.41	313.15	0.337	719.44	343.15	34.990	731.90	378.14	34.968	703.32	413.13	34.965	674.40	448.13	34.975	644.82
278.20	34.968	784.84	313.15	4.977	725.70	348.14	0.529	680.38	383.14	0.803	636.88	418.13	1.304	586.18	453.13	2.113	520.24
278.21	39.953	788.06	313.16	9.978	731.86	348.15	4.974	688.70	383.13	4.979	648.35	418.13	4.954	602.47	453.13	4.969	545.61
283.21	0.360	750.94	313.15	14.971	737.52	348.15	9.970	696.95	383.13	9.981	659.84	418.13	9.961	619.40	453.13	9.964	573.55
283.21	4.974	755.80	313.15	19.967	742.72	348.15	14.970	704.31	383.13	14.975	669.63	418.13	14.960	632.83	453.13	14.987	592.82
283.21	9.970	760.69	313.16	24.983	747.57	348.14	19.984	710.98	383.13	19.974	678.21	418.13	19.974	644.12	453.12	19.987	607.83
283.21	14.979	765.25	313.16	29.982	752.06	348.14	24.973	717.05	383.13	24.975	685.90	418.13	24.980	653.88	453.13	24.983	620.29
283.21	19.959	769.49	313.16	34.966	756.29	348.14	29.972	722.66	383.13	29.988	692.81	418.13	29.993	662.47	453.12	29.981	631.00
283.21	24.972	773.48	313.15	39.549	760.02	348.15	34.977	727.83	383.13	34.978	699.18	418.13	34.974	670.24	453.12	34.987	640.41
283.21	29.976	777.23	318.16	0.416	714.15	353.14	0.557	674.48	388.13	0.859	630.17	423.13	1.393	578.02	458.12	2.257	508.00
283.21	34.981	780.76	318.16	4.966	720.58	353.14	4.965	683.16	388.13	4.985	642.23	423.13	4.980	595.38	458.13	4.983	536.00
283.21	39.971	784.07	318.16	9.962	726.99	353.14	9.978	691.82	388.13	9.981	654.30	423.13	9.986	613.36	458.13	9.963	566.12
288.21	0.366	745.81	318.16	14.982	732.87	353.15	14.972	699.46	388.13	14.979	664.52	423.13	14.980	627.42	458.13	14.972	586.48
288.21	4.963	750.86	318.16	19.966	738.24	353.15	19.978	706.36	388.13	19.980	673.44	423.13	19.975	639.11	458.12	19.968	602.17
288.21	9.986	755.96	318.16	24.978	743.25	353.14	24.979	712.65	388.13	24.974	681.37	423.13	24.978	649.18	458.13	24.980	615.14
288.21	14.967	760.66	318.16	29.992	747.89	353.14	29.969	718.42	388.13	29.972	688.51	423.13	29.973	658.11	458.13	29.972	626.21
288.21	19.971	765.06	318.16	34.967	752.24	353.14	34.985	723.75	388.13	34.978	695.07	423.13	34.960	666.06	458.12	34.989	635.89
288.21	24.982	769.18	323.16	0.429	708.69	358.14	0.594	668.49	393.13	0.920	623.30	428.13	1.495	569.62	463.12	2.409	494.29
288.21	29.980	773.05	323.16	4.976	715.41	358.14	4.986	677.60	393.13	4.960	635.89	428.13	4.957	587.85	463.12	4.951	525.01
288.21	34.972	776.67	323.16	9.978	722.09	358.14	9.985	686.63	393.13	9.976	648.66	428.13	9.979	607.11	463.13	9.956	558.28
288.21	39.970	780.10	323.16	14.972	728.16	358.14	14.976	694.58	393.13	14.986	659.36	428.13	14.981	621.89	463.13	14.989	579.98
293.21	0.368	740.64	323.16	19.967	733.74	358.14	19.963	701.71	393.13	19.984	668.63	428.13	19.984	634.08	463.13	19.987	596.40
293.21	4.959	745.89	323.15	24.987	738.92	358.14	24.982	708.23	393.13	24.964	676.81	428.13	24.965	644.49	463.13	24.983	609.83
293.21	9.968	751.18	323.16	29.983	743.71	358.14	29.978	714.17	393.13	29.972	684.20	428.13	29.980	653.71	463.13	29.983	621.24
293.21	14.964	756.06	323.16	34.971	748.18	358.14	34.984	719.66	393.13	34.974	690.94	428.13	34.988	661.87	463.13	34.978	631.23
293.21	19.966	760.61	328.15	0.446	703.18	363.14	0.623	662.38	398.13	0.980	616.24	433.13	1.602	560.97	468.12	2.575	478.56
293.21	24.974	764.87	328.15	4.968	710.17	363.14	4.964	671.89	398.13	4.976	629.55	433.13	4.964	580.21	468.13	4.978	513.42
293.21	29.986	768.86	328.15	9.964	717.12	363.14	9.983	681.38	398.13	9.966	642.96	433.13	9.977	600.74	468.13	9.977	550.13
293.21	34.962	772.60	328.15	14.983	723.46	363.14	14.968	689.65	398.13	14.982	654.15	433.13	14.981	616.28	468.12	14.970	573.05
293.21	39.959	776.12	328.15	19.992	729.24	363.14	19.959	697.05	398.13	19.992	663.78	433.13	19.982	628.98	468.13	19.985	590.33
298.22	0.378	735.43	328.15	24.969	734.55	363.14	24.983	703.79	398.13	24.974	672.27	433.13	24.971	639.77	468.12	24.984	604.30
298.22	4.968	740.90	328.15	29.985	739.52	363.14	29.989	709.93	398.13	29.971	679.90	433.13	29.984	649.24	468.12	29.986	616.10
298.22	9.966	746.38	328.15	34.973	744.12	363.14	34.963	715.59	398.13	34.963	686.82	433.13	34.973	657.67	468.12	34.973	626.39
298.21	14.968	751.45	333.15	0.463	697.59	368.14	0.661	656.18	403.13	1.051	609.03	438.13	1.722	551.39	473.12	2.748	459.11
298.22	19.983	756.17	333.15	4.959	704.87	368.14	4.962	666.12	403.13	4.953	622.96	438.13	4.972	572.28	473.12	4.981	500.38
298.22	24.979	760.55	333.15	9.977	712.15	368.14	9.963	676.04	403.13	9.968	637.20	438.13	9.972	594.22	473.12	9.953	541.33
298.22	29.969	764.66	333.15	14.978	718.71	368.14	14.968	684.69	403.13	14.967	648.88	438.13	14.988	610.61	473.12	14.955	565.85
298.22	34.979	768.53	333.15	19.970	724.68	368.14	19.979	692.40	403.13	19.979	658.92	438.13	19.994	623.84	473.12	19.986	584.03
298.22	39.949	772.14	333.15	24.982	730.21	368.14	24.977	699.34	403.13	24.970	667.69	438.13	24.969	634.99	473.13	24.977	598.60
303.22	0.387	730.18	333.15	30.000	735.31	368.14	29.978	705.67	403.13	29.979	675.57	438.13	29.967	644.76	473.12	29.976	610.87
303.22	4.972	735.88	333.15	34.972	740.05	368.14	34.975	711.48	403.13	34.973	682.68	438.13	34.980	653.43	473.13	34.964	621.46
303.22	9.976	741.58															

been measured by Govender et al.[11] (for MTBE and DIPE from 288 K up to 328 K and up to 8 MPa), Jangkamolkulchai et al.[12] (for MTBE and ETBE from 273 K up to 333 K and up to 0.8 MPa), Schornack and Eckert[13] (for DIPE from 303 K up to 323 K and at pressures from 107 MPa up to 511 MPa), Senger[14] (for MTBE and ETBE from 298 K up to 328 K and up to 207 MPa), and Ulbig et al.[15] (for DIPE from 278 K up to 323 K and up to 60 MPa). All published density data for the liquid phase were measured below or near the normal boiling points. Therefore, the new measurements represent an enormous extension of the $P\rho T$ data available for MTBE, ETBE, and DIPE.

The measured densities were correlated with the three-dimensional $P\rho T$-correlating model TRIDEN.[3–5] In this model the Tait equation for isothermal compressed densities was combined with a modified Rackett equation for the liquid saturation densities and the Wagner vapor pressure equation in the "2.5,5" form, used as a reference state (ρ_0 and P_0), which is required for the Tait equation. Using these equations, it is possible to correlate the $P\rho T$ data in the whole liquid state up to the critical point, nearly within experimental error. The required parameters for the Wagner equation (eq 1) are taken from the *Dortmund Data Bank for Pure Component Properties*.[10] These parameters were fitted using experimental vapor pressure data from different researchers.

$$\ln(P_0/\mathrm{kPa}) = \ln(P_c/\mathrm{kPa}) + \frac{A_W(1 - T_r) + B_W(1 - T_r)^{1.5} + C_W(1 - T_r)^{2.5} + D_W(1 - T_r)^5}{T_r} \quad (1)$$

At temperatures below the normal boiling point, the reference pressure is set to $P_0 = 0.1013$ MPa. The reference

Table 2. Experimental Densities of Ethyl *tert*-Butyl Ether (ETBE)

T/K	P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}	T/K	P/MPa	ρ/kg·m^{-3}
273.21	0.295	760.69	303.22	39.962	768.22	338.15	34.970	736.59	378.13	0.597	646.81	408.12	29.968	674.17	443.12	14.981	611.99
273.21	4.968	765.26	308.22	0.367	725.12	338.14	39.965	741.11	378.13	4.959	657.54	408.12	34.998	681.24	443.12	19.955	624.70
273.21	9.955	769.80	308.22	4.982	731.04	343.14	0.440	687.56	378.13	9.985	668.06	408.12	39.962	687.77	443.12	24.981	635.64
273.21	14.970	774.06	308.22	9.958	736.92	343.14	4.974	695.45	378.13	14.956	677.08	413.12	0.922	600.90	443.12	29.975	645.17
273.21	19.979	778.04	308.22	14.976	742.36	343.14	9.959	703.17	378.13	19.966	685.13	413.12	4.971	616.03	443.12	34.969	653.67
273.21	24.962	781.78	308.22	19.965	747.37	343.14	14.977	710.16	378.12	24.977	692.40	413.12	9.976	630.72	443.12	39.976	661.34
273.21	29.967	785.31	308.22	24.965	752.03	343.14	19.983	716.49	378.13	29.978	698.88	413.12	14.964	642.72	448.12	1.534	546.34
273.21	34.980	788.63	308.22	29.977	756.40	343.14	24.968	722.27	378.12	34.979	704.96	413.12	19.984	653.03	448.12	4.960	568.37
273.21	39.969	791.78	308.22	34.956	760.48	343.14	29.977	727.64	378.13	39.961	710.53	413.12	24.976	662.02	448.12	9.956	590.25
278.21	0.353	755.73	308.22	39.980	764.33	343.14	34.963	732.61	383.13	0.634	640.62	413.12	29.968	670.05	448.12	14.954	606.60
278.21	4.967	760.41	313.15	0.379	719.91	348.14	0.457	681.96	383.13	4.953	651.83	413.12	34.992	677.34	448.12	19.970	619.87
278.21	9.981	765.15	313.16	4.968	726.05	348.14	4.973	690.19	383.13	9.967	662.80	413.12	39.963	684.00	448.12	24.966	631.14
278.21	14.972	769.54	313.16	9.983	732.19	348.14	9.958	698.24	383.13	14.979	672.23	418.12	0.994	593.81	448.12	29.976	640.97
278.21	19.966	773.65	313.15	14.954	737.77	348.14	14.975	705.49	383.13	19.970	680.55	418.12	4.953	609.65	448.11	34.979	649.67
278.21	24.977	777.53	313.16	19.973	742.96	348.14	19.984	712.04	383.13	24.988	688.03	418.12	9.964	625.16	448.12	39.965	657.54
278.21	29.971	781.15	313.16	24.969	747.77	348.14	24.974	718.01	383.13	29.968	694.77	418.12	14.968	637.72	453.12	1.651	537.37
278.21	34.966	784.58	313.16	29.982	752.26	348.14	29.960	723.52	383.13	34.953	700.97	418.12	19.985	648.44	453.11	4.970	560.79
278.21	39.968	787.82	313.16	34.961	756.47	348.14	34.965	728.65	383.13	39.981	706.70	418.11	24.978	657.73	453.12	9.953	584.04
283.21	0.354	750.71	313.16	39.973	760.42	353.14	0.478	676.30	388.13	0.672	634.34	418.12	29.972	665.99	453.12	14.965	601.23
283.21	4.980	755.58	318.15	0.386	714.66	353.14	4.983	684.91	388.13	4.977	646.14	418.12	34.981	673.48	453.11	19.967	615.00
283.21	9.976	760.47	318.16	4.958	721.05	353.14	9.958	693.28	388.13	9.957	657.56	418.12	39.974	680.27	453.11	24.967	626.61
283.21	14.981	765.04	318.16	9.959	727.40	353.14	14.987	700.81	388.13	14.967	667.39	423.11	1.062	586.53	453.12	29.975	636.71
283.21	19.966	769.27	318.16	14.964	733.22	353.14	19.968	707.56	388.13	19.964	675.99	423.12	4.961	603.24	453.12	34.983	645.66
283.21	24.987	773.27	318.16	19.965	738.58	353.14	24.979	713.73	388.13	24.973	683.70	423.11	9.954	619.55	453.11	39.978	653.69
283.21	29.968	777.01	318.15	24.984	743.56	353.14	29.965	719.41	388.13	29.970	690.67	423.12	14.965	632.68	458.12	1.775	527.84
283.21	34.980	780.55	318.16	29.971	748.17	353.14	34.966	724.69	388.13	34.997	697.03	423.12	19.978	643.74	458.12	4.961	552.76
283.21	39.976	783.88	318.16	34.972	752.51	358.14	0.497	670.55	388.13	39.968	702.92	423.12	24.981	653.34	458.11	9.955	577.66
288.21	0.356	745.65	318.16	39.969	756.56	358.14	4.956	679.51	393.13	0.712	627.91	423.11	29.974	661.84	458.12	14.964	595.69
288.21	4.964	750.70	323.15	0.374	709.31	358.14	9.962	688.29	393.13	4.973	640.33	423.12	34.975	669.52	458.11	19.963	609.99
288.21	9.955	755.77	323.15	4.955	715.99	358.14	14.974	696.09	393.13	9.975	652.34	423.12	39.968	676.50	458.11	24.956	621.97
288.21	14.959	760.49	323.15	9.957	722.61	358.14	19.966	703.09	393.13	14.960	662.52	428.11	1.143	579.03	458.11	29.973	632.39
288.21	19.961	764.89	323.15	14.963	728.63	358.14	24.972	709.46	393.13	19.971	671.45	428.11	4.971	596.67	458.12	34.961	641.55
288.21	24.981	769.01	323.15	19.975	734.18	358.14	29.978	715.33	393.12	24.956	679.37	428.12	9.957	613.88	458.11	39.979	649.79
288.21	29.976	772.88	323.15	24.983	739.31	358.14	34.980	720.74	393.12	29.978	686.58	428.12	14.969	627.58	463.11	1.910	517.64
288.21	34.967	776.51	323.15	29.967	744.06	363.13	0.520	664.74	393.12	34.958	693.10	428.11	19.978	639.05	463.11	4.970	543.86
288.21	39.975	779.94	323.15	34.980	748.53	363.14	4.960	674.11	393.12	39.977	699.13	428.11	24.984	648.96	463.12	9.957	571.04
293.21	0.354	740.57	323.15	39.968	752.68	363.13	9.961	683.27	398.12	0.762	621.40	428.11	29.985	657.70	463.11	14.965	589.98
293.21	4.961	745.82	328.15	0.402	703.98	363.14	14.985	691.39	398.12	4.951	634.36	428.11	34.968	665.55	463.11	19.961	604.86
293.21	9.962	751.09	328.15	4.955	710.91	363.14	19.980	698.62	398.12	9.976	647.04	428.11	39.959	672.73	463.11	24.971	617.24
293.21	14.978	755.98	328.15	9.984	717.82	363.13	24.978	705.18	398.12	14.965	657.64	433.11	1.226	571.32	463.11	29.977	627.96
293.21	19.958	760.51	328.15	14.965	724.04	363.13	29.980	711.21	398.12	19.974	666.89	433.12	4.963	589.83	463.11	34.970	637.35
293.21	24.968	764.76	328.15	19.961	729.76	363.14	34.979	716.79	398.12	24.974	675.08	433.12	9.958	608.09	463.11	39.979	645.80
293.21	29.965	768.75	328.15	24.967	735.05	368.13	0.523	658.79	398.12	29.984	682.46	433.12	14.970	622.40	468.11	2.061	506.58
293.21	34.958	772.49	328.15	29.972	739.96	368.13	4.972	668.67	398.12	34.951	689.17	433.12	19.980	634.28	468.12	4.979	535.00
293.21	39.969	776.03	328.15	34.970	744.55	368.13	9.967	678.20	398.12	39.970	695.37	433.12	24.990	644.51	468.11	9.956	564.07
298.21	0.364	735.46	328.15	39.981	748.82	368.14	14.960	686.60	403.12	0.807	614.72	433.12	29.983	653.50	468.11	14.958	584.00
298.21	4.965	740.92	333.15	0.414	698.57	368.13	19.976	694.13	403.12	4.954	628.36	433.12	34.983	661.60	468.11	19.968	599.54
298.22	9.971	746.39	333.15	4.952	705.79	368.13	24.968	700.88	403.12	9.974	641.67	433.12	39.965	668.90	468.11	24.959	612.34
298.21	14.977	751.45	333.15	9.979	712.97	368.13	29.967	707.11	403.12	14.977	652.74	438.12	1.323	563.39	468.11	29.968	623.36
298.22	19.961	756.13	333.15	14.974	719.43	368.13	34.988	712.84	403.12	19.980	662.31	438.12	4.966	582.89	468.11	34.967	633.02
298.22	24.971	760.52	333.15	19.971	725.35	368.13	39.968	718.12	403.12	24.965	670.74	438.12	9.961	602.24	468.11	39.980	641.67
298.21	29.981	764.62	333.15	24.970	730.80	373.13	0.570	652.88	403.12	29.969	678.34	438.12	14.973	617.21	473.11	2.217	494.21
298.22	34.986	768.49	333.15	29.977	735.86	373.14	4.962	663.14	403.12	34.997	685.22	438.12	19.960	629.49	473.11	4.963	524.94
298.21	39.970	772.12	333.15	34.965	740.57	373.13	9.973	673.17	403.12	39.971	691.59	438.12	24.980	640.08	473.11	9.963	556.68
303.22	0.368	730.31	333.15	39.975	744.97	373.13	14.989	681.91	408.12	0.865	607.87	438.12	29.983	649.34	473.11	14.957	577.71
303.22	4.978	736.00	338.15	0.427	693.09	373.13	19.967	689.63	408.12	4.961	622.21	438.12	34.980	657.65	473.11	19.967	593.92
303.22	9.973	741.68	338.15	4.964	700.64	373.13	24.974	696.64	408.12	9.985	636.22	438.12	39.970	665.13	473.11	24.958	607.20
303.22	14.984	746.92	338.15	9.973	708.09	373.13	29.982	703.03	408.12	14.966	647.71	443.12	1.421	554.75	473.11	29.966	618.56
303.21	19.970	751.76	338.14	14.969	714.80	373.13	34.956	708.89	408.12	19.983	657.65	443.12	4.980	575.83	473.11	34.975	628.51
303.21	24.966	756.27	338.14	19.973	720.92	373.13	39.964	714.34	408.12	24.969	666.35	443.12	9.959	596.30	473.11	39.968	637.37
303.22	29.971	760.51	338.15	24.979	726.54												
303.22	34.988	764.49	338.14	29.974	731.75												

densities ρ_0 at P_0 (saturation pressure or 0.1013 MPa) are extrapolated from the compressed liquid density measurements and correlated with the modified Rackett equation:

$$\rho_0 = A_R / B_R^{[1+(1-T/C_R)^{D_R}]} \quad (2)$$

For the Tait equation

$$\rho = \rho_0 \Big/ \left[1 - C_T \ln\left(\frac{B_T + P}{B_T + P_0}\right)\right] \quad (3)$$

the following temperature dependence is used for the parameter B_T:

$$B_T = b_0 + b_1 \frac{T}{E_T} + b_2\left(\frac{T}{E_T}\right)^2 + b_3\left(\frac{T}{E_T}\right)^3 \quad (4)$$

And for the parameter C_T a linear temperature dependence is used:

$$C_T = c_0 + c_1\left(\frac{T}{E_T}\right) \quad (5)$$

Besides a deviation plot, other statistical values are desirable to evaluate the correlation. The absolute (RMSD)

Table 3. Experimental Densities of Diisopropyl Ether (DIPE)

T	P	ρ	T	P	ρ	T	P	ρ	T	P	ρ	T	P	ρ	T	P	ρ
K	MPa	$kg\cdot m^{-3}$	K	MPa	$kg\cdot m^{-3}$	K	MPa	$kg\cdot m^{-3}$	K	MPa	$kg\cdot m^{-3}$	K	MPa	$kg\cdot m^{-3}$	K	MPa	$kg\cdot m^{-3}$
273.21	0.340	744.30	303.22	34.977	749.65	338.15	24.983	711.31	373.14	19.960	674.01	408.13	14.984	631.54	443.13	9.968	578.82
273.21	4.975	749.13	303.22	39.909	753.53	338.15	29.976	716.77	373.14	24.979	681.41	408.13	19.996	642.04	443.13	14.979	595.66
273.21	9.967	753.97	308.22	0.374	708.22	338.15	34.973	721.82	373.14	29.979	688.10	408.13	24.978	651.21	443.13	19.976	609.19
273.21	14.977	758.48	308.22	4.976	714.57	343.15	0.453	670.06	373.14	34.952	694.22	408.13	29.985	659.41	443.13	24.979	620.64
273.21	19.972	762.69	308.22	9.966	720.84	343.15	4.962	678.55	378.14	0.624	628.31	408.13	34.953	666.76	443.13	29.969	630.60
273.21	24.967	766.63	308.22	14.957	726.57	343.15	9.956	686.84	378.14	4.980	640.08	413.13	0.993	580.90	443.13	34.967	639.43
273.21	29.989	770.34	308.22	19.973	731.88	343.15	14.966	694.25	378.14	9.986	651.37	413.13	4.956	597.53	448.13	1.619	523.39
273.21	34.976	773.83	308.22	24.967	736.79	343.15	19.971	700.94	378.14	14.983	661.03	413.13	9.967	613.59	448.13	4.962	548.53
278.20	0.349	739.27	308.22	29.981	741.38	343.15	24.972	707.05	378.14	19.981	669.52	413.13	14.980	626.51	448.13	9.979	572.81
278.21	4.980	744.28	308.22	34.987	745.64	343.15	29.985	712.68	378.14	24.987	677.10	413.13	19.982	637.40	448.13	14.984	590.40
278.21	9.961	749.28	313.15	0.356	702.88	343.15	34.941	717.87	378.14	29.979	684.02	413.13	24.971	646.85	448.13	19.986	604.46
278.21	14.966	753.95	313.16	4.967	709.52	348.14	0.471	664.36	378.14	34.959	690.29	413.13	29.973	655.28	448.13	24.981	616.26
278.21	19.984	758.30	313.16	9.980	716.05	348.15	4.979	673.26	383.14	0.672	621.97	413.13	34.944	662.84	448.13	29.986	626.51
278.20	24.977	762.38	313.16	14.972	721.99	348.15	9.962	681.88	383.14	4.961	634.25	418.13	1.061	573.45	448.13	34.980	635.53
278.21	29.993	766.20	313.16	19.984	727.47	348.15	14.957	689.55	383.14	9.959	646.08	418.13	4.952	590.99	453.13	1.732	513.86
278.20	34.952	769.78	313.16	24.976	732.53	348.15	19.984	696.50	383.14	14.980	656.18	418.13	9.970	607.98	453.13	4.969	540.90
283.21	0.348	734.18	313.16	29.976	737.23	348.15	24.981	702.79	383.14	19.992	665.00	418.13	14.966	621.42	453.13	9.975	566.65
283.21	4.977	739.39	313.16	34.968	741.63	348.15	29.985	708.57	383.14	24.969	672.80	418.13	19.964	632.70	453.13	14.955	585.01
283.21	9.969	744.59	318.16	0.392	697.60	348.15	34.967	713.91	383.14	29.951	679.88	418.13	24.973	642.50	453.13	19.978	599.66
283.21	14.981	749.42	318.16	4.985	704.50	353.14	0.492	658.57	383.14	34.957	686.36	418.13	29.969	651.17	453.13	24.975	611.84
283.21	19.968	753.90	318.16	9.968	711.25	353.15	4.951	667.83	388.14	0.711	615.48	418.13	34.968	658.92	453.13	29.977	622.38
283.21	24.976	758.11	318.16	14.961	717.40	353.15	9.971	676.89	388.14	4.968	628.40	423.13	1.143	565.83	453.13	34.984	631.64
283.21	29.976	762.05	318.16	19.989	723.07	353.15	14.973	684.87	388.14	9.962	640.80	423.13	4.990	584.48	458.13	1.865	504.02
283.21	34.969	765.75	318.16	24.984	728.30	353.15	19.979	692.02	388.14	14.981	651.31	423.13	9.967	602.28	458.13	4.980	532.49
288.21	0.358	729.07	318.16	29.976	733.15	353.15	24.970	698.52	388.14	19.967	660.40	423.13	14.992	616.39	458.13	9.980	560.43
288.21	4.965	734.47	318.16	34.964	737.67	353.15	29.984	704.47	388.14	24.976	668.51	423.13	19.961	628.02	458.13	14.957	579.66
288.21	9.977	739.88	323.15	0.400	692.21	353.15	34.968	709.97	388.14	29.985	675.82	423.13	24.971	638.14	458.13	19.973	594.83
288.21	14.960	744.86	323.16	4.983	699.39	358.14	0.505	652.69	388.14	34.947	682.42	423.13	29.968	647.05	458.13	24.962	607.41
288.21	19.960	749.50	323.15	9.957	706.41	358.14	4.965	662.43	393.14	0.759	608.88	423.13	34.978	655.01	458.13	29.989	618.26
288.21	24.977	753.85	323.15	14.972	712.82	358.14	9.987	671.89	393.14	4.984	622.49	428.13	1.225	557.98	458.13	34.984	627.77
288.21	29.990	757.92	323.16	19.973	718.66	358.14	14.982	680.16	393.14	9.971	635.48	428.13	4.960	577.55	463.13	1.999	493.60
288.21	34.977	761.71	323.15	24.967	724.05	358.14	19.970	687.54	393.14	14.992	646.41	428.13	9.970	596.54	463.13	4.954	524.03
293.21	0.363	723.92	323.16	29.971	729.05	358.14	24.981	694.26	393.13	19.959	655.80	428.13	14.966	611.20	463.13	9.963	554.02
293.21	4.977	729.54	323.16	34.963	733.69	358.14	29.959	700.38	393.14	24.972	664.18	428.13	19.970	623.35	463.13	14.960	574.28
293.21	9.961	735.13	328.15	0.413	686.76	358.14	34.947	706.02	393.14	29.975	671.70	428.13	24.975	633.77	463.13	19.956	589.99
293.21	14.960	740.30	328.15	4.981	694.25	363.14	0.534	646.76	393.13	34.954	678.50	428.13	29.981	642.94	463.13	24.968	603.01
293.21	19.972	745.11	328.15	9.976	701.59	363.14	4.956	656.91	398.13	0.810	602.12	428.13	34.959	651.12	463.13	29.971	614.15
293.21	24.979	749.59	328.15	14.984	708.22	363.14	9.966	666.78	398.13	4.957	616.36	433.13	1.311	549.63	463.13	34.970	623.91
293.21	29.971	753.77	328.15	19.969	714.24	363.14	14.961	675.38	398.14	9.971	630.09	433.13	4.968	570.61	468.13	2.140	482.60
293.21	34.962	757.70	328.15	24.980	719.82	363.14	19.981	683.06	398.14	14.956	641.43	433.13	9.969	590.71	468.13	4.961	515.39
298.22	0.368	718.73	328.15	29.980	724.95	363.14	24.978	689.98	398.13	19.963	651.22	433.13	14.965	606.04	468.13	9.968	547.62
298.22	4.981	724.58	328.15	34.966	729.73	363.14	29.981	696.27	398.13	24.980	659.88	433.13	19.971	618.65	468.13	14.960	568.84
298.22	9.961	730.38	333.15	0.425	681.27	363.14	34.971	702.09	398.13	29.965	667.59	433.13	24.973	629.40	468.13	19.960	585.16
298.22	14.967	735.74	333.15	4.980	689.08	368.14	0.564	640.71	398.13	34.953	674.58	433.13	29.979	638.84	468.13	24.974	598.60
298.22	19.974	740.71	333.15	9.977	696.71	368.14	4.959	651.35	403.13	0.867	595.22	433.13	34.961	647.22	468.13	29.974	610.04
298.22	24.981	745.32	333.15	14.982	703.59	368.14	9.956	661.65	403.13	4.973	610.24	438.13	1.405	541.16	468.13	34.968	620.03
298.22	29.971	749.62	333.15	19.965	709.81	368.14	14.981	670.64	403.13	9.983	624.69	438.13	4.980	563.52	473.13	2.291	470.93
298.22	34.971	753.67	333.15	24.978	715.57	368.14	19.977	678.56	403.13	14.968	636.49	438.13	9.975	584.83	473.13	4.964	506.31
303.22	0.369	713.49	333.15	29.985	720.86	368.14	24.980	685.70	403.13	19.974	646.64	438.13	14.973	600.86	473.13	9.978	541.12
303.22	4.966	719.57	333.15	34.964	725.78	368.14	29.993	692.19	403.13	24.972	655.53	438.13	19.973	613.92	473.13	14.966	563.37
303.22	9.969	725.62	338.15	0.439	675.70	368.14	34.969	698.15	403.13	29.966	663.49	438.13	24.980	625.03	473.13	19.964	580.31
303.22	14.967	731.16	338.15	4.954	683.81	373.14	0.600	634.58	403.13	34.942	670.67	438.13	29.963	634.70	473.13	24.982	594.17
303.22	19.963	736.29	338.15	9.985	691.82	373.14	4.962	645.73	408.13	0.923	588.13	438.13	34.976	643.32	473.13	29.987	605.90
303.22	24.965	741.05	338.15	14.978	698.93	373.14	9.969	656.53	408.13	4.959	603.93	443.13	1.510	532.48	473.13	34.965	616.14
303.22	29.981	745.49	338.15	19.970	705.38	373.14	14.963	665.81	408.13	9.959	619.13	443.13	4.954	556.03			

and relative (RMSDr) root-mean-square deviations and the mean deviation (bias) are used as statistical values for the TRIDEN fits.

$$\mathrm{RMSD} = \sqrt{\frac{1}{n}\sum_{n}(\rho_{\mathrm{exp}} - \rho_{\mathrm{calc}})^2} \qquad (6)$$

$$\mathrm{RMSDr} = 100\sqrt{\frac{1}{n}\sum_{n}\left(\frac{\rho_{\mathrm{exp}} - \rho_{\mathrm{calc}}}{\rho_{\mathrm{exp}}}\right)^2} \qquad (7)$$

$$\mathrm{bias} = \frac{1}{n}\sum_{n}(\rho_{\mathrm{exp}} - \rho_{\mathrm{calc}}) \qquad (8)$$

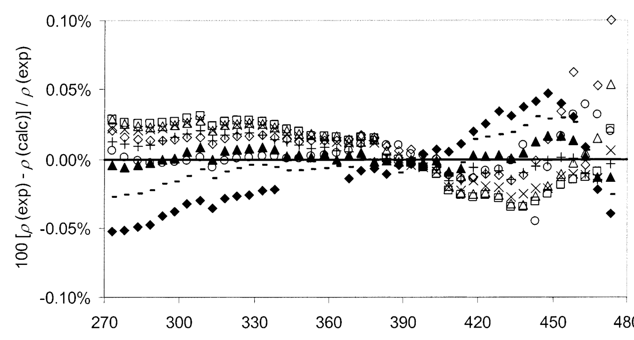

Figure 6. Relative deviations between experimental densities and the TRIDEN correlation for ethyl *tert*-butyl ether (ETBE): ○, <5 MPa; ◇, 5 MPa; △, 10 MPa; □, 15 MPa; ×, 20 MPa; +, 25 MPa; ▲, 30 MPa; −, 35 MPa; ◆, 40 MPa.

The TRIDEN parameters for the Tait equation, the Rackett equation, and the Wagner equation, the temperature and pressure range covered, and additional statistical values are given in Table 4.

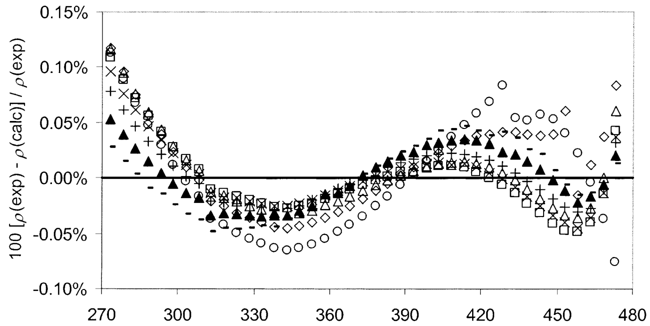

Figure 7. Relative deviations between experimental densities and the TRIDEN correlation for diisopropyl ether (DIPE): ○, <5 MPa; ◇, 5 MPa; △, 10 MPa; □, 15 MPa; ×, 20 MPa; +, 25 MPa; ▲, 30 MPa; −, 35 MPa.

Table 4. Parameters for the TRIDEN Correlation Model for Methyl *tert*-Butyl Ether (MTBE), Ethyl *tert*-Butyl Ether (ETBE), and Diisopropyl Ether (DIPE): Temperature Range, Pressure Range, Number of Data Points, Tait Parameters, Rackett Parameters, Wagner Parameters, Critical Temperature, Critical Pressure, Normal Boiling Point, Absolute (RMSD) and Relative (RMSDr) Root-Mean-Square Deviations, and the Mean Deviation (bias) as Statistical Values for the TRIDEN Fit

	MTBE	ETBE	DIPE
T_{min}/K	273.2	273.2	273.2
T_{max}/K	473.1	473.1	473.1
P_{min}/MPa	0.32	0.29	0.34
P_{max}/MPa	40	40	35
ρ_{min}/kg·m^{-3}	459.1	494.2	470.9
ρ_{max}/kg·m^{-3}	792.1	791.8	773.8
no. of data points	337	364	328
c_0	0.079 805 6	0.082 464 6	0.082 860 8
c_1	0	-1.4587×10^{-6}	0
b_0/MPa	408.936	404.898	317.385
b_1/MPa	−212.638	−208.134 64	−146.968
b_2/MPa	37.7063	36.6551	21.7937
b_3/MPa	−2.344 75	−2.267 32	−1.039 11
E_T/K	100	100	100
A_R/kg·m^{-3}	141.206	161.444	52.1000
B_R	0.376 705	0.401 211	0.232 377
C_R/K	478.081	486.318	505.577
D_R	0.379 821	0.437 625	0.253 450
RMSD/kg·m^{-3}	0.2813	0.1314	0.2421
RMSDr/%	0.0478	0.0194	0.0353
bias/kg·m^{-3}	0.0014	0.0166	0.0138
A_W	−9.004 54	−7.834 36	−7.362 13
B_W	6.979 72	3.456 17	1.918 67
C_W	−10.3256	−7.127 94	−5.893 25
D_W	9.921 31	7.182 01	12.1373
T_c/K	497.1	509.4	500.0
P_c/kPa	3434.9	2933.9	2877.6
T_b/K	328.2	345.5	341.5

In Figures 5−7 the relative deviations between experimental values and the correlation are shown. The deviations are usually within ±0.1% except at the extreme temperatures. Comparisons were also made between the TRIDEN correlation and the mentioned high-pressure densities published by other researchers. In Figures 8−10 the relative deviations between published values and the TRIDEN correlation are shown for MTBE, ETBE, and DIPE. The values of Jangkamolkulchai et al.[12] for MTBE and ETBE and of Ulbig et al.[15] for DIPE are in good agreement with the correlations with relative root-mean-square deviations of 0.06%, 0.05%, and 0.17%, respectively. Also, the densities for MTBE and ETBE measured by Senger[14] up to 40 MPa show good agreement with the correlations (RMSDr: 0.18% for both), with larger deviations obtained with rising pressure (up to +0.5% at 298 K and 40 MPa for both ethers). Larger deviations are observed for the values of Govender et al.[11] for MTBE and DIPE with RMSDr values of 0.86% and 0.60%, respectively.

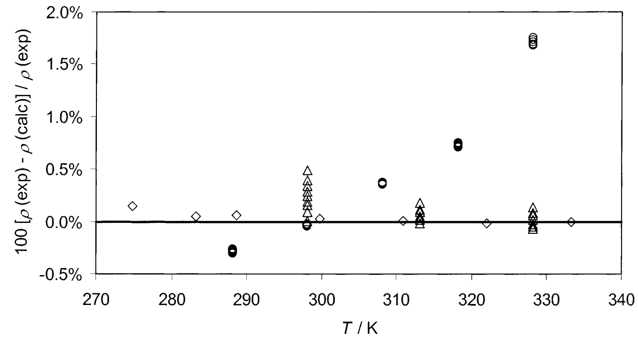

Figure 8. Relative deviations between published densities and the TRIDEN correlation for methyl *tert*-butyl ether (MTBE): ○, ref 11; ◇, ref 12; △, ref 14.

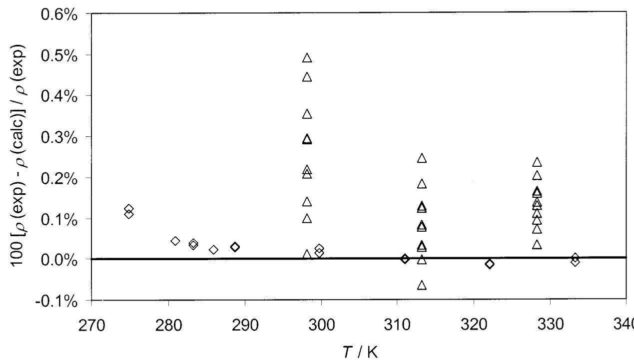

Figure 9. Relative deviations between published densities and the TRIDEN correlation for ethyl *tert*-butyl ether (ETBE): ◇, ref 12; △, ref 14.

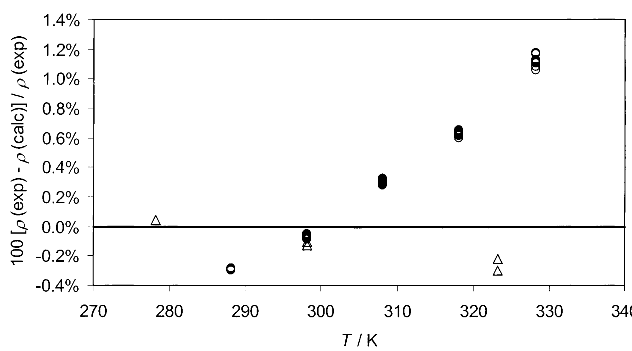

Figure 10. Relative deviations between published densities and the TRIDEN correlation for diisopropyl ether (DIPE): ○, ref 11; △, ref 15.

With rising temperatures the densities of Govender et al. show increasing deviations for MTBE and DIPE up to +1.5% and +1.3% (at 328 K), respectively.

The relative root-mean-square deviation between the DDB−Pure correlation[10] (from literature values of different researchers) for saturated liquid densities and the calculation with TRIDEN (extrapolation to the saturation pressure) is 0.27%, 0.04%, and 0.11% for MTBE (273 K to 458 K), ETBE (273 K to 308 K), and DIPE (273 K to 333 K), respectively.

Summary and Outlook

Densities in the compressed liquid state are presented for methyl *tert*-butyl ether (MTBE), ethyl *tert*-butyl ether (ETBE), and diisopropyl ether (DIPE) for temperatures between 273 K and 473 K and pressures up to 40 MPa. These measurements represent a wide extension of the $P\rho T$ data available for MTBE, ETBE, and DIPE. All data were correlated with the TRIDEN model and show good agreement with data published by other authors.

The density measurements for MTBE, ETBE, and DIPE are a continuation of the density measurements already performed for toluene, carbon dioxide, carbonyl sulfide, hydrogen sulfide, sulfur hexafluoride, dinitrogen monoxide, and R227ea.[3,4,6] In the future, densities in the compressed liquid phase for γ-butyrolactone and *N*-methyl-2-pyrrolidone (NMP) will be published. Furthermore, measurements on compressed liquid densities and excess volumes for binary mixtures will be presented.

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